

Development of Laser Induced Capillary Wave Method for Viscosity Measurement Using Pulsed Carbon Dioxide Laser¹

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We have developed a new experimental apparatus based on laser induced capillary wave method by using pulsed carbon dioxide laser (wavelength 10.6 μm , pulse width 50 ns, power 65 mJ) as a heating source. Since the present technique is applicable to wide range of the viscosity, it is possible to satisfy the needs in the food industry. In this method, the interfered laser beams heat a liquid surface and they generate capillary wave (wavelength can be adjusted from 20 to 200 μm) caused by spatially sinusoidal temperature distribution. The temporal behavior of capillary wave is detected by a diffracted probe beam (He-Ne laser, 15 mW) at the heated area. The dynamics of the capillary wave provides information on the thermophysical properties such as the viscosity and the surface tension. In the present study, we have measured several liquid samples (e.g. acetone, toluene, ethanol, 1-hexanol, ethylene glycol and glycerol) at room temperature. The detected signal for low viscosity liquid agrees well with the theoretical calculation taking into account the influence of the surface tension distribution.

KEY WORDS: laser induced capillary wave; measurement technique; surface tension; viscosity.

1. INTRODUCTION

The viscosity is one of the very important thermophysical properties for food industry, because the viscosity is essential in the design and control of production processes and is also useful for evaluation of the food quality. Moreover the viscosity of food material

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considerably changes in the food processing. Therefore it is required to develop a new technique which is possible to measure significantly changing the viscosity by single apparatus. However, non-contact technique for the viscosity measurement applicable to wide range of the viscosity has not been available, so the conventional measurement techniques such as capillary viscometer (for low viscous liquid) and rotational viscometer (for high viscous liquid), etc. are used for the viscosity measurement of the fluid foods. In the present study, we have developed a new method to detect the capillary wave under the transient grating geometry. We called this technique “laser induced capillary wave method”.

The measurement techniques which provides information on the viscosity and the surface tension by observing the spontaneous capillary wave (ripplon) caused by the thermal fluctuations has been reported, e.g. Surface Laser-Light Scattering (SLLS, Nishio *et al.* [1]) and Surface Light Scattering (Fröba *et al.* [2]). However the experimental techniques SLLS have difficulty in the case of measuring highly viscous liquid because the spontaneous capillary wave is over damped. In the meantime, Terazima *et al.* [3] has reported the motion of the capillary wave in a manner similar to that of the present method. The one of main differences between Terazima’s experimental apparatus and the present one is the heating source. Terazima employs the Nd:YAG laser, but on the other hand we employs the CO₂ laser. The CO₂ laser has several advantages over Nd:YAG laser, for example, it is less necessary to add the absorbing materials in the samples and is possible to design more flexible optical system because of its high coherency.

In the present paper, we describe the principle of measurement and experimental apparatus by using pulsed carbon dioxide laser and discuss the comparison of detected signals with theoretical calculations for several liquid samples having wide variety of the viscosity.

2. THE PRINCIPLE OF MEASUREMENT

2.1. Creation and Detection of Laser Induced Capillary Wave

The principle of laser induced capillary wave technique is shown schematically in Fig. 1. In this method, pulsed high-power laser beams of equal wavelength and equal intensity intersect on a sample surface under a crossing angle θ . They generate an optical interference fringe pattern as illustrated in Fig. 1, whose intensity distribution is spatially sinusoidal. The laser intensity distribution and wave number can be written as

$$I(x) = I_h \{1 + \cos(kx)\} \quad (1)$$

$$k = \frac{2\pi}{\Lambda} = \frac{2\sin(\theta/2)}{\lambda_h} \quad (2)$$

where I_h is the heating laser beam intensity, Λ the fringe space, λ_h the wavelength of

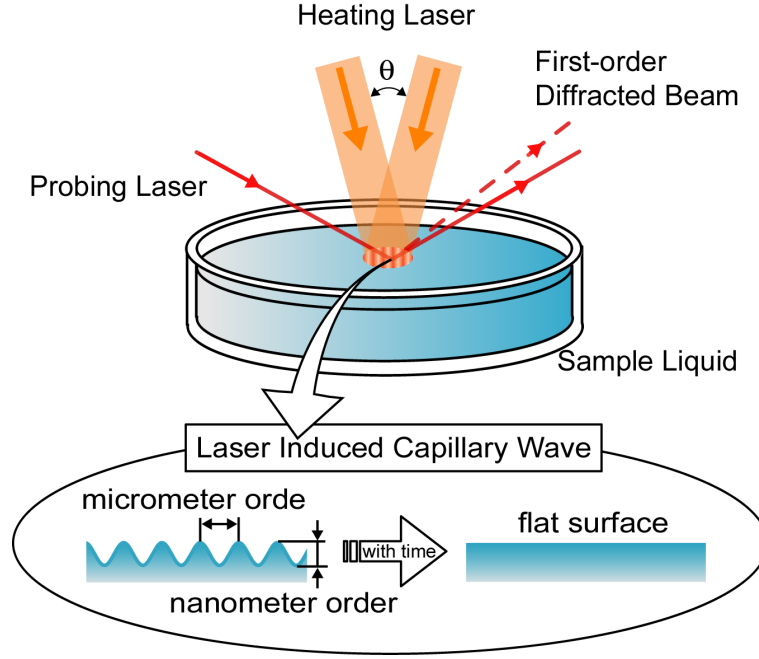


Fig. 1 The principle of the laser induced capillary method. The laser induced capillary wave has micrometer order wavelength and nanometer order amplitude.

heating laser. At the end of heating, a corresponding spatially sinusoidal temperature distribution induced by the grating pattern is

$$T(x) = T_m + \Delta T \cos(kx) \quad (3)$$

Where T_m is the mean initial temperature rise, ΔT is the amplitude of spatially periodic temperature distribution.

The temperature distribution of the thermal grating creates capillary wave on the liquid surface driven by the thermal expansion and the temperature dependence of the surface tension. The displacement of capillary wave along z direction can be reasonably assumed by

$$u_z = u_m + \Delta u \cos(kx) \quad (4)$$

here u_m is spatially uniform displacement and Δu is the amplitude of laser induced capillary wave. In the present experimental set-up, the amplitude of created capillary wave is estimated to be about 10nm. When the probing laser beam is incident on the heating area, the laser beam is diffracted because the capillary waves act like the reflection grating. The first-order diffraction efficiency is directly proportional to the square of the surface displacement as the following equation:

$$I(t) \propto \Delta u(t)^2 \quad (5)$$

where $I(t)$ is intensity of the first-order diffraction beam. Therefore we are able to obtain the information of the behavior of laser induced capillary wave by measuring the

time profile of the first-order diffraction beam. Particularly, the temporal behavior of laser induced capillary wave is described in the next sections (2.2. and 2.3.). Besides, we employ short-pulsed CO₂ laser in the present technique in order to neglect the complex motion of laser induced capillary wave in the course of heating process.

2.2. The Motion of Laser Induced Capillary Wave

The motion of laser induced capillary wave is described by three fundamental equations under several boundary conditions. The displacement of the Newtonian liquid surface has been reported by Terajima *et al.* [3] and is summarized here in after.

First, the Navier Stokes equation is

$$\frac{\partial^2 \mathbf{u}}{\partial t^2} - \nu \nabla^2 \frac{\partial \mathbf{u}}{\partial t} - V_L \nabla \nabla \cdot \mathbf{u} = -\beta V_L^2 \nabla T \quad (6)$$

where \mathbf{u} is the displacement, β the thermal expansion coefficient, V_L the sound velocity in the liquid and $\nu = \eta / \rho$ the kinetic viscosity (η ; the viscosity, ρ ; the density of the medium). Here we assume that the displacement of the capillary wave on the surface is smaller than the wavelength enough to consider as laser induced capillary wave is equal to the spontaneous capillary wave. In addition, the convective term is neglected. Second, the continuity equation is expressed as

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \mathbf{V}) = 0 \quad (7)$$

where \mathbf{V} is the velocity of the liquid. Third, the equation of heat conduction can be obtained by the following equation:

$$\rho C_p \left(\frac{\partial T}{\partial t} \right) - \lambda \nabla^2 T = I_h \alpha [1 + \cos(kx)] \exp(\alpha z) \delta(t) \quad (8)$$

Here λ is the thermal conductivity, α the optical penetration length of the heating beam, C_p the heat capacity at a constant pressure and $\delta(t)$ Dirac delta function.

The boundary conditions are as follows, the velocity of liquid should be zero at sufficiently far from the surface and the thermal conduction from the liquid to the gas phase is neglected. In addition, the tangential and normal components of the stress to the surface, which is assumed under the conditions that the stress acting from within the liquid is exactly counter balanced by the stress due to the surface force since no stress exist in the gas phase, is give by

$$\sigma \frac{\partial u_x}{\partial x^2} - 2 \frac{\partial}{\partial t} \left(\frac{\partial u_z}{\partial z} \right) + \rho V_L^2 \left(\alpha T - \frac{\partial u_x}{\partial x} - \frac{\partial u_z}{\partial z} \right) = 0 \quad (9)$$

$$\eta \frac{\partial}{\partial t} \left(\frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x} \right) - \left(\frac{\partial \sigma}{\partial T} \right) \frac{\partial T}{\partial x} = 0 \quad (10)$$

Especially, the temperature dependence of the surface tension is considered by using Eq.

(10) in which the surface tension distribution term is included.

The amplitude of the laser induced capillary wave can be provided by solving Eqs. (4) and (6)-(10) using Laplace transformation. The Laplace transformed amplitude of the wave ΔU can be written as

$$\Delta U = \frac{\alpha\beta}{\lambda k^2} \frac{I_e}{(b_2^2 - b_1^2)} \times \left[\frac{b_1 b_5}{1 + b_5} \left\{ -\frac{e^{b_1 k z}}{b_4^2 - b_1^2} + \frac{e^{b_2 k z}}{b_4^2 - b_2^2} \right\} - \frac{b_2 - b_1}{b_2} (C_1 e^{b_3 k z} + b_4 C_2 e^{b_4 k z}) \right] \quad (11)$$

where

$$b_1 = \alpha/k \quad (12)$$

$$b_2 = (1 + s/ak^2)^{1/2} \quad (13)$$

$$b_3 = (1 + s/vk^2)^{1/2} \quad (14)$$

$$b_4 = \left[1 + \frac{s^2}{(V_L^2 + s v)k^2} \right] \quad (15)$$

$$b_5 = \frac{V_L^2}{s v} \quad (16)$$

and both C_1 and C_2 are extracted by solving determinant:

$$\begin{bmatrix} 2b_3 + d_1 b_5 & 2b_4^2 + b_5(b_4^2 + d_1 b_4 - 1) \\ 1 + b_3^2 & 2b_4 \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \end{bmatrix} = \begin{bmatrix} d_2 d_3 - b_5 \\ 2b_1 b_2 (b_1 + b_2) d_3 - \frac{\partial \sigma}{\partial T} \frac{k}{\beta \rho V_L^2} b_5 \end{bmatrix} \quad (17)$$

Here

$$d_1 = \frac{\sigma k}{\rho V_L^2} \quad (18)$$

$$d_2 = b_1 b_2 \left\{ (b_5 + 2)(b_4^2 + b_1 b_2) + d_1 b_5 (b_1 + b_2) \right\} - b_5 (b_1^2 + b_1 b_2 + b_2^2 - b_4^2) \quad (19)$$

$$d_3 = \frac{b_5}{(1+b_5)(b_4^2 - b_1^2)(b_4^2 - b_2^2)} \quad (20)$$

The time profile of the liquid motion is numerically calculated from the Eq. (11) by using the fast inverse Laplace transform algorithm [4].

2.3. The Mechanism of Laser induced Capillary Wave Generation

In the early stage of our study, we assumed that the motion of laser induced capillary wave is similar to that of ripplon [5-7]. However we found the influence of the temperature dependence of the surface tension is extremely significant under the present experimental conditions. Since the laser induced capillary wave is generated by the temperature distribution, the surface tension distribution corresponding to the temperature distribution is caused simultaneously. That is to say, it is necessary to consider the influence of the temperature dependence of the surface tension and we apply the Eq. (10) to one of the boundary conditions. The temperature dependence of the surface tension is one of the important factors in relation to the behavior of laser induced capillary wave.

Figure 2 shows the amplitude of the laser induced capillary wave on a surface of toluene as illustration of low viscous liquid by calculation of Eq. (11) using the fast inverse Laplace transform. The main thermophysical properties of liquids which we have used in theoretical calculations are listed in Table I. The (a) wave is calculated under the assumption which is no temperature dependence of the surface tension on the surface. In calculation of (b), it is assumed that the thermal expansion does not exist. The sum of (a) and (b) is equal to the theoretical amplitude of laser induced capillary wave, and (b) is the dominant motion of laser induced capillary wave, i.e. the temperature dependence of the surface tension is one of the significant factors for the wave generation. Accordingly, we have demonstrated that the temperature dependence of the surface

Table I. Thermophysical Properties of Liquids Used in Theoretical Calculation (at 298 K and 1 atm)[8].

		Toluene	Glycerol
η	(mPa·s)	0.552	945
σ	(mN·m ⁻¹)	27.3	63.4 (293 K)
$d\sigma/dT$	(mN·m ⁻¹ ·K ⁻¹)	-0.118 (283-373 K) [9]	-0.0537 (293-373 K) [10]
ρ	(kg·m ⁻³)	862	1263
β	(10 ⁻⁶ ·K ⁻¹)	1060 (273-303 K)	490 (288-303 K)
C_p	(kJ·kg ⁻¹ ·K ⁻¹)	1.717	2.46
λ	(mW·m ⁻¹ ·K ⁻¹)	133	287

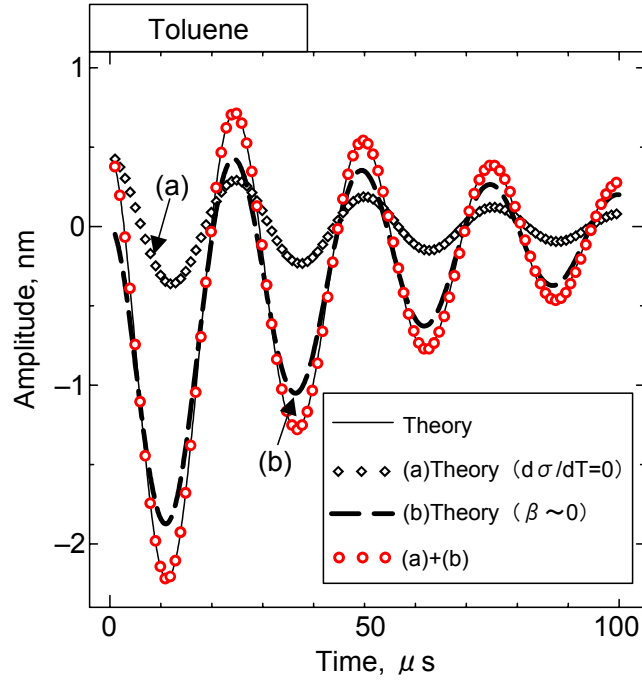


Fig. 2 The theoretical calculation of amplitude of laser induced capillary wave versus time for toluene.

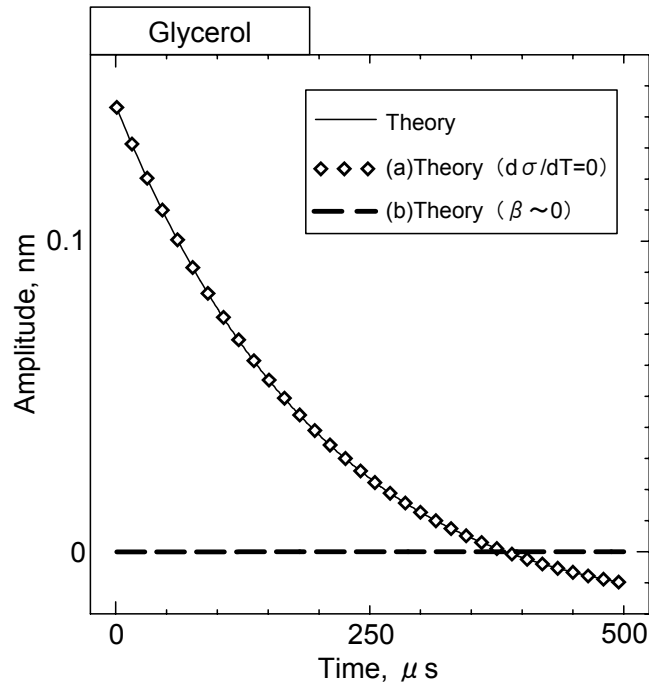


Fig. 3 The theoretical calculation of amplitude of laser induced capillary wave versus time for glycerol.

tension is the essential contribution for the wave generation in addition to the thermal expansion under the present experimental conditions. However the dominant contribution for the wave generation on high viscous liquid is not the temperature dependence of the surface tension but the thermal expansion. We show the amplitude of laser induced capillary wave on a surface of glycerol in Fig. 3, where each wave of (a)

and (b) is calculated under the assumption as same as the case of calculation for toluene. As can be seen in Fig. 3, the theoretical amplitude of laser induced capillary wave for glycerol is in good agreement with (a). Specifically, the generation of laser induced capillary wave on the high viscous liquid is caused by only thermal expansion.

3. EXPERIMENTAL APPARATUS

Figure 4 exhibits the present experimental apparatus. The pulsed CO₂ laser (Edinburgh Instruments Ltd., MTL-3; wavelength 10.6 μm , pulse width 50 ns, output energy 65 mJ) is employed as a heating source. The repetition rates are available from single shot to 100Hz. Especially, because of the short-pulsed laser beam, it is possible to neglect the complex behavior of laser induced capillary wave in the course of heating process. The pulsed heating beam is turned vertically up by mirror 1 (M1) and divided into two beams of equal intensity by means of a beam splitter (BS). They are intersected on the sample surface by refracting in M2, M3 and M4 to produce an interference pattern. These optical devices are fixed on the vertical optical bench and the radius of the heating beams is approximately 6 mm on the sample surface. Typically the range of fringe space of the grating is adjusted from about 20 to 200 μm . The liquid sample is filled in a petri dish (depth 1.6mm, radius 5 cm). The probing laser is a He-Ne laser (wavelength 632.8 nm, output power 15 mW, beam radius 2 mm). The diffracted light signal as a function of time is detected by a photomultiplier tube (PMT; Hamamatsu, R928) through a pinhole (PH) and an interference filter (IF). The output signal is sent to a digital storage oscilloscope (DSO) and is transferred to a computer. The entire apparatus is arranged on an optical bench.

In order to determine the fringe space prior to the experiment, the CO₂ laser

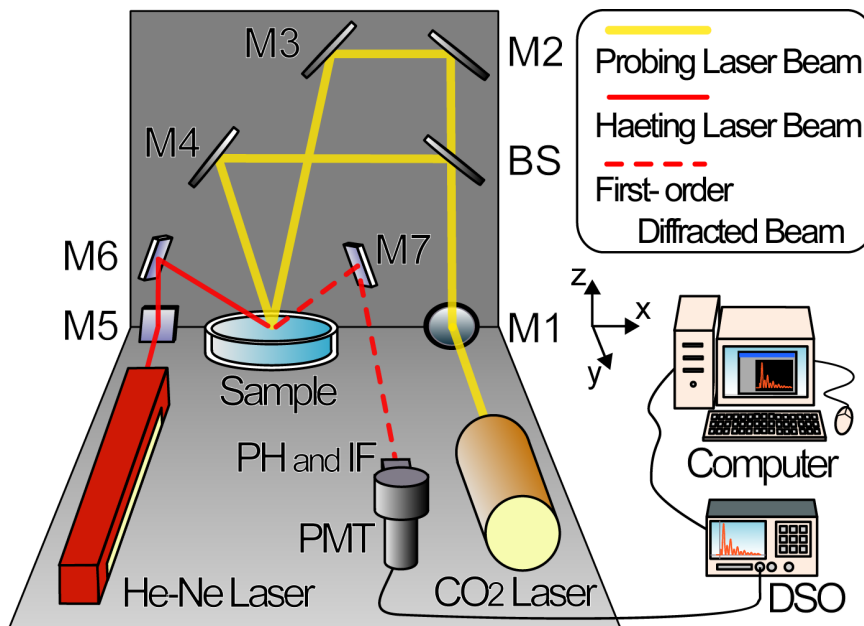


Fig. 4 The experimental apparatus of laser induced capillary method by using pulsed CO₂ laser.

beams intersect on the surface of the transparent PMMA plate (thickness 2 mm), which set at the same position as the sample surface. Consequently, the grating pattern which is generated by the same optical configuration as the experiment is printed on the surface of the PMMA plate. When the He-Ne laser is incident on the printed grating, the diffraction beam is generated and we can obtain the grating period by measuring the diffraction angle.

Laser induced capillary wave method has the characteristics in that it is possible to measure the viscosity (1) of various liquids which have wide range of the viscosity, (2) at high speed, (3) in contact-free manner because of using optical system and (4) without additional absorbing materials in the samples by using pulsed CO₂ laser. Thus, laser induced capillary wave method is a quite unique technique.

4. EXPERIMENTAL RESULTS AND DISCUSSION

As the first step, in order to confirm the principle of the present method, we have employed pure liquids such as acetone, toluene, ethanol, 1-hexanol, ethylene glycol and glycerol.

As an example, we show the detected signal of the present technique for toluene (viscosity is 0.55 mPa·s at 298 K), which was measured under the condition at room temperature (approximately 298 K), atmospheric pressure and wavelength of capillary wave (equal to fringe space of heating beams) 50 μm , and the theoretical waveform, which is calculated by Eqs. (5) and (11) using thermophysical properties listed in Table I,

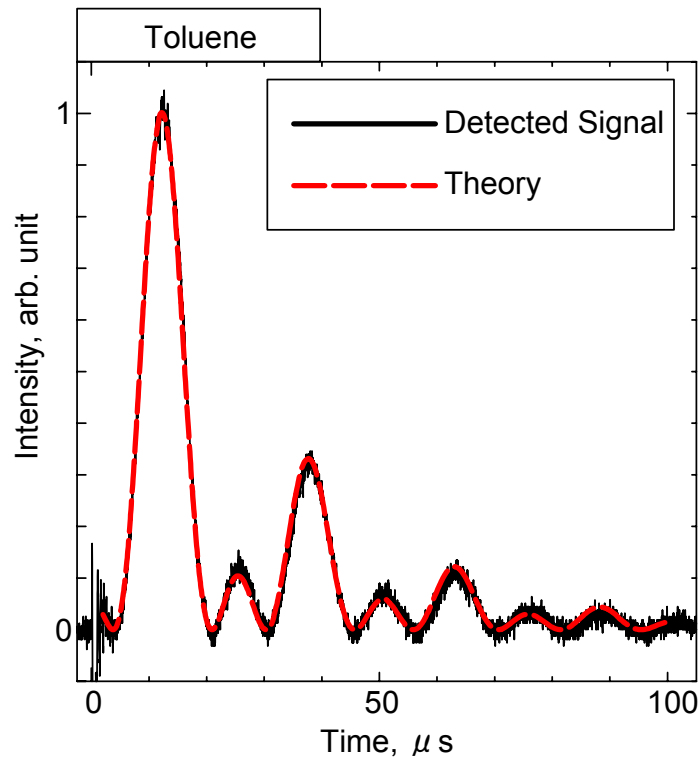


Fig. 5 Comparison of the detected signal with the theoretical calculation for toluene at room temperature.

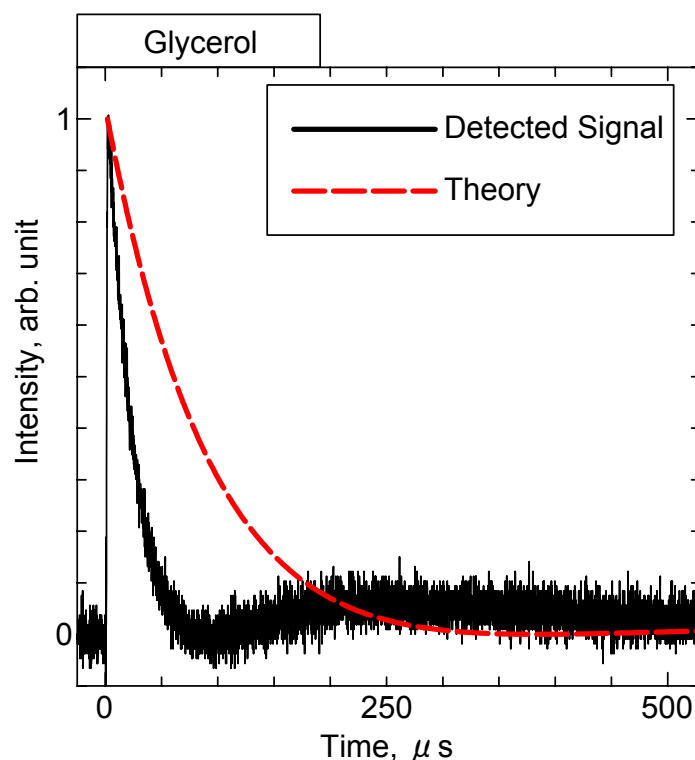


Fig. 6 Comparison of the detected signal with the theoretical calculation for glycerol at room temperature.

in Fig. 5. Here each of results is normalized at the highest peak. In the case of measuring toluene which is one of the typical low viscous liquids, the detected signal is damping oscillation wave. The detected signal for toluene agrees well with the theoretical calculation which was taken into account the effect of the surface tension distribution. On the other hand, the detected signal shown in Fig. 6 is over damping, when we measure the high viscosity sample such as glycerol (viscosity is 945 mPa·s at 298K) under the same conditions as for toluene. Although we have the detected signals agreeable to the theory for toluene which have low viscosity, the signal for glycerol which is one of the highest viscous liquids in the measured samples is not in agreement with theory completely as for toluene, but each of the detected signal and theory is over damping waveform. Thus we have obtained the characteristic detected signals for each of toluene and glycerol in comparison with Figs. 5 and 6. These remarkably different waveforms can be explained by the difference of viscosity, i.e. the viscosity of glycerol is over 1000 times as that of toluene.

Additionally, the comparisons of the detected signals with the theoretical calculations for acetone (viscosity is 0.30 mPa·s at 298K), ethanol (viscosity is 1.1 mPa·s at 298 K), 1-hexanol (viscosity is 4.6 mPa·s at 298K) and ethylene glycol (viscosity is 14 mPa·s at 298K) are shown in Fig. 7 (a)-(d) in the viscosity order. The agreement between the theory and the detected signals is not only confirmed by toluene but also by acetone, ethanol, 1-hexanol and ethylene glycol. Beside the detected signals for acetone, ethanol, 1-hexanol and ethylene glycol are the characteristic waveforms as well as for

toluene and glycerol in comparison with Figs. 5, 6 and 7. Consequently, we have obtained the detected signals according to the viscosity of several samples which conform to the theoretical calculation by laser induced capillary wave method.

The present study leads to the conclusions that the laser induced capillary wave method has the great possibility for measuring the wide range of the viscosity, which requires neither contact with the sample nor absorbable materials in the samples by using pulsed CO₂ laser, and the mechanism of laser induced capillary wave generation consists two contributions of thermal expansion and the temperature dependence of the surface tension. In the near future, it is necessary to derive the inverse problem to determine the viscosity by the detected signal.

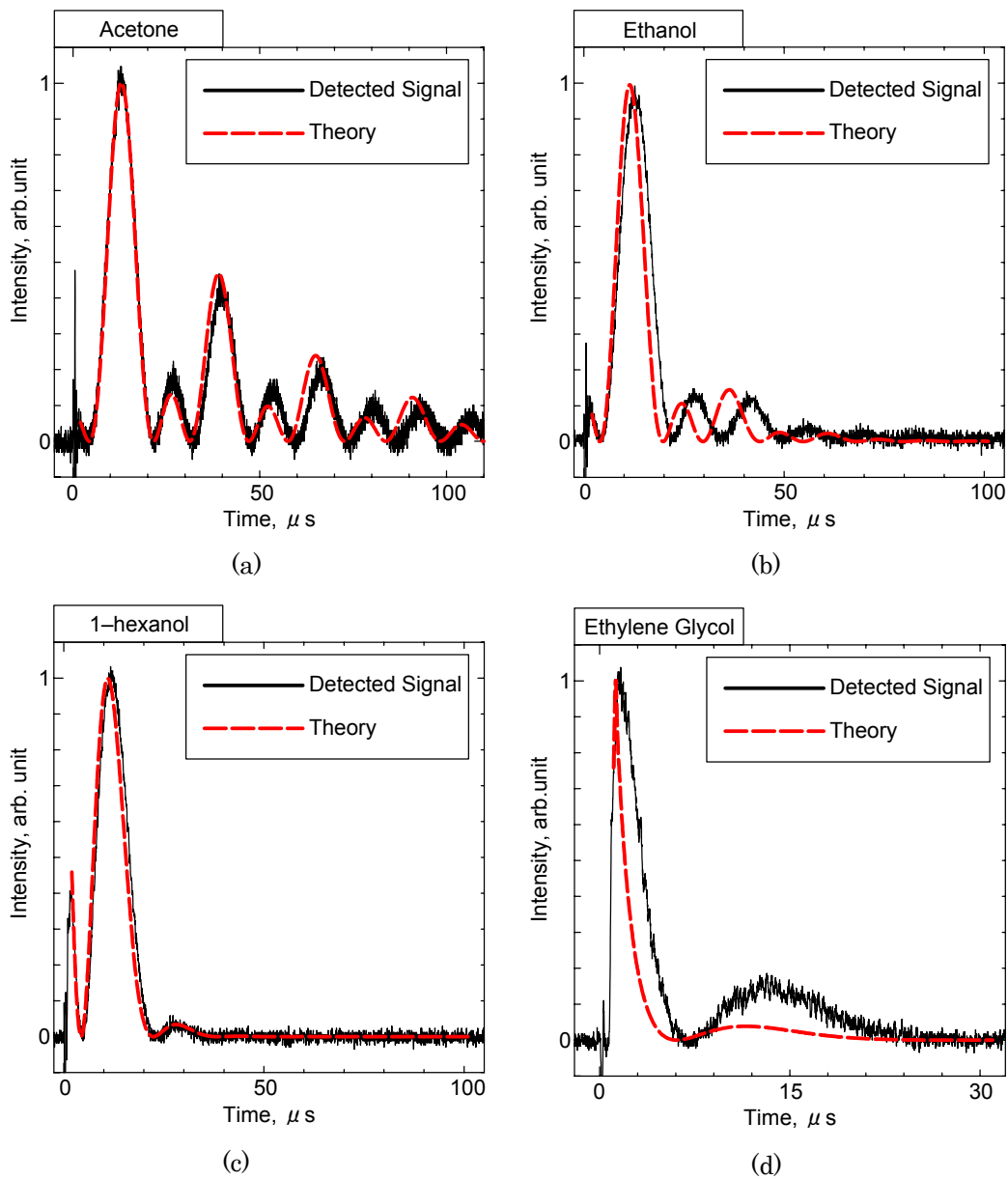


Fig. 7 Comparison of the detected signal with the theoretical calculation for (a) acetone (b) ethanol (c) 1-hexanol (d) ethylene glycol at room temperature.

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